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Charge Trap Assisted High Efficiency in New Polymer-Blend Based Light Emitting Diodes

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Abstract

Polymer blend system, $F8_{1-x}SY_x$, based on mixture of poly (9,9-dioctylfluorene) (F8) and a poly (*para*-phenylenevinylene) (PPV) copolymer, superyellow (SY), has been proven to be a high performance blend material in polymer light emitting diode (PLED). This blend system exhibits luminance (L) > 10^4 cd-m^{-2}, luminous efficiency (η) > 21 cd-A^{-1} and low operating voltage (V) ~ 3-10 V. The performance can be credited to the large difference (~ 0.6 eV) between the highest occupied molecular orbital (HOMO) levels of F8 (~ 5.8 eV) and SY (~ 5.2 eV), where, SY molecules serve as hole-traps in the F8 host polymer and reduce their mobility. This dictates a balanced charge injection into the emissive layer and results in overall increase in the device performance.

Organic semiconductors have been attracting immense attention for last two decades because of their unique features including easy tunability, viable processing, flexibility and excellent optoelectronic properties.[1-10] Device applications, such as polymer light-emitting diodes (PLEDs), photovoltaic cells and organic thin-film field-effect transistor (OTFTs) have already found their way towards commercialization.[11-15] With this immense success, further improvement in their performance is naturally a consistent goal. To achieve this, a range of strategies have been adopted. For example, in the field of PLEDs, making layered structures and using thick emissive layers have been proven very useful in increasing the device performance.[16-20] Using polymer blends as emissive layer is another powerful tool in order to enhance the efficiency as well tune the emission wavelength.[21, 22] Various electrode schemes have also been exploited which allow efficient and balance charge injection in the emissive layer, an effective technique that has achieved the same goal.[23]

Multiple emissive layers in polymer heterostructure devices are fabricated in layer-by-layer fashion, such that each layer uses a cross-linker as bonder which reduces the miscibility between the adjacent layers.[8, 24] However, partial dissolution of the pre-deposited layer usually occurs with the deposition of the subsequent layer resulting roughness and film retention issues at the interface. Thermal layer stabilization and using orthogonal solvents have also been used to fabricate layered structure giving off blue emission.[25] Thick PLEDs (with emissive layer thickness of ~ 1 μm) are attractive as they are easy to fabricate and show high luminous efficiency.[19, 20] However, they require very large operating voltage, limiting their use in many on-chip circuit applications. Such devices also consume more material. Using polymer-blends in PLEDs offers its own advantages because, a) fabrication of the emissive layer involves only one deposition step of the emissive layer and mixing of adjacent emissive layers is not applicable in contrast to heterostructure light emitting devices, b) these

devices require low operating voltage, and c) very high efficiency can be achieved in blend based PLEDs (as reported here).

In the past, primary focus for blend based PLEDs remained on systems such as poly-fluorene (PF) blends, e.g. poly(9,9-dioctylfluorene) (F8) + poly(9,9-dioctylfluorene-*alt*-benzothiadiazole) (F8BT) [26-29], poly(9,9-dioctylfluorene-*alt*-N-(4-butylphenyl)-diphenylamine) (TFB) + F8BT,[30, 31] and PFs + PPVs blends, e.g. poly[2-methoxy-5-(2′-ethyl-hexyloxy)-*p*-phenylenevinylene] (MEH-PPV) doped in F8.[32, 33] In this report, we purpose a polymer-polymer blend-system, $F8_{1-x}SY_x$ (where x signifies the weight fraction of SY in F8), a mixture of F8 and a PPV-copolymer known as superyellow (SY). F8 is an attractive polymer due to its efficient blue photoluminescence (PL) and electroluminescence (EL).[29, 34, 35] It also serves as a backbone host in many white light-emitting copolymers because its emission can be tuned easily by copolymerization with different complimentary comonomers.[29] SY offers efficient and stable performance and long operational lifetime (100,000 hrs). [36-39] It also offers much higher PL quantum efficiency than other PPV derivatives,[37] making it a favorable material for light emitting devices. We chose to make this system firstly, because of strong spectral overlap between emission spectra of F8 and absorption spectra of SY – there is a potential of strong energy transfer between these molecules via Förster resonance energy transfer. Secondly, different charge transport properties of both polymers offer us an opportunity to see whether a balanced charge transport can be achieved within the emissive layer that could eventually lead to efficient devices.

We present our experimental findings for $F8_{1-x}SY_x$ based PLEDs as a function of increasing SY weight concentration, x. This blend system exhibits remarkable properties: luminance (L) > 10^4 cd-m^{-2}, luminous efficiency (η) > 21 cd-A^{-1} and, low operating voltage (V) ~ 3-

10 V. We believe that the working principle lies in the large difference (~ 0.6 eV) between the highest occupied molecular orbital (HOMO) levels of F8 (~ 5.8 eV) and SY (~ 5.2 eV). [40-42] SY molecules serve as hole-traps in the F8 host polymer and reduce their mobility. This dictates the charge balance / injection into the blend emissive layer and results in overall increase in the device performance. In order to understand the charge transport in $F8_{1-x}SY_x$ system, hole-only and electron-only devices for different values of *x* were also made. We use Mott-Gurney space-charge-limited current (SCLC) equation combined with field-dependent mobility[43, 44] in order to estimate the hole mobility. Our findings support the hypothesis we made before this study that modified hole transport (due to SY traps) is responsible for very high efficiency of $F8_{1-x}SY_x$ based PLEDs. We anticipate that the system can pave the way for different device applications because of its superior electrical and optical properties as compared with the other well-known polymer blend systems. To the best of our knowledge, this is the first report for utilizing F8 and SY in a blend form for light emitting applications.

Two types of devices (bipolar and unipolar) were fabricated for different weight concentrations of SY, $x = 0$, 0.05, 0.1, 0.25, 0.5, 0.95 and 1, respectively. Electro-emissive properties were studied in a layered configuration of bipolar PLEDs (glass/ITO/PEDOT:PSS/$F8_{1-x}SY_x$/Ca/Al). Schematic representation of the bipolar device along with the chemical structure and respective energy band diagram (emissive layer highlighted) are shown in **Figure 1**. Charge transport properties were studied in unipolar devices, subcategorized as hole-only (glass/ITO/PEDOT:PSS/$F8_{1-x}SY_x$/MoO_3/Au) and electron-only device (Glass/ITO/ZnO/Cs_2CO_3/$F8_{1-x}SY_x$/Ca/Al) in order to gain a deeper insight for understanding the charge transport within the emissive layer.

Figure 2a describes the *J-V* characteristics of bipolar devices for different values of *x*. Device with $x = 0$ (pure F8) exhibits its maximum current density (J_{max}) ≈ 1.7×10^4 mA-cm^{-2} at a safe operating voltage of ~ 5 V. This is the highest value of the current density at this operating

voltage across the whole range of concentrations. Subsequently, there wasis a dramatic fall in current density with mixing of merely a small amount of SY (x = 0.05) in F8, which indicates the severe charge-trapping nature of SY molecules in F8 matrix.[45] However, for the following concentrations we observe an increasing trend in current density (under identical operating voltages) which persisted throughout the entire voltage sweep for all subsequently increasing values of x. It was noticed that J-V curves follow a systematic trend with increasing value of x. F8 loses its trend only at x = 0.05 – the composition roughly exhibits a mixed trend of F8 and SY, however, with a value of current density that is three orders of magnitude smaller than pure SY. Whereas all higher concentrations made within $0.05 < x < 1$ exhibit a trend resembling more like that for x = 1 (pure SY). This indicates that the charge transport is mainly dominated by SY molecules in the $F8_{1-x}$:SY_x system. We observed that the turn-on voltage (V_{on}) behavior was also systematic with changing SY concentration: there was little difference in V_{on} ($\approx$ 2.4 V) between x = 0 and x = 0.05. However, a gradual decrease is noticed with increasing SY across the entire range of $0.05 < x \leq 1$, with a minimum of value of $V_{on} \approx$ 2 V obtained for x = 1, see **Table 1** for details.

The luminance vs. voltage (L-V) curves for the range $0.05 \leq x \leq 1$ almost follow J-V trends, **Figure 2b**. The maximum values of luminance for x = 0, 0.05 and 1 are ~ 2.5×10^2 (at 5 V), 2.5×10^4 (at 10 V) and 4.6×10^4 cd-m^{-2} (at 10 V), respectively. We again noticed that the inclusion of merely a small amount of SY in F8 causes significant changes: L_{max} improves by nearly 2 orders of magnitude and devices withstand much larger voltages compared with pure F8 devices. All PLEDs within $0.05 \leq x \leq 1$ can be operated safely up to 10 V. The luminous-efficiency vs. voltage (η-V) curves for increasing value of x exhibit an interesting picture, **Figure 2c**. Pure F8 and SY devices exhibit their respective highest efficiencies $\eta \approx 0.9$ (at ~ 3.4 V) and ~ 14.2 cd-A^{-1} (at ~ 5.4 V), respectively. PLED with x = 0.05 shows the highest efficiency, $\eta \approx 21.2$ (at ~ 4.4 V) – about 23 (1.5) times higher than pure F8(SY) PLED. The

second highest value is 20.6 cd-A^{-1} (at ~ 4.4 V) for 0.1 (see **Supporting Information S1** for power efficiency and external quantum efficiency for $x = 0.05$ and 0.1 concentrations). This is a significant improvement as compared with the devices which are based only on the single constituting polymer, F8 and SY. [37, 40, 46, 47]

We believe that the improved performance of $F8_{1-x}SY_x$ PLEDs is because of the efficient charge balance mediated by SY hole-traps. Hole mobility in F8 is orders of magnitude larger than the electron mobility.[46] Therefore, most of the emission is expected to occur near to the cathode surface, resulting in low luminance efficiency for pure F8 devices. HOMO level of SY is at ~ 5.2 eV, which when mixed in F8, resulted in hole traps because of the large difference between HOMO levels of both molecules (~ 0.6 eV).[41] These traps strongly suppress the hole mobility. Since LUMOs of F8 and SY are at the same level,[36, 41] presumably resulting in a lesser change in electron transport (compared with pure F8), at least for small SY concentration such as $x = 0.05$ and 0.1. Consequently, a charge balance is achieved – emission zone is pushed away from the cathode surface and device efficiency is improved. In order to understand the charge transport in $F8_{1-x}SY_x$ blend system, we also studied single charge carrier devices (discussed later).

Comparative study of the PL and EL spectra of PLEDs help understanding their luminescence properties. Both spectra normalized to their respective highest peaks for $F8_{1-x}SY_x$ within $0 \leq x \leq 1$ are given in **Figure 3a,b,** respectively. In general, there is a consistency between the PL and the EL spectra of blend and pure SY PLEDs. However, we observed that the PL spectrum of pure F8 exhibits less green emission at ~ 530 nm compared to its EL spectrum. The underlying reason is that the PL involves the optical excitation of the thin film followed by the recombination from the excited states, whereas, EL results from the recombination of injected charges during their transport across the biased film. Therefore, a smaller electron injection barrier can be the reason for this higher EL efficiency. The origin

of green emission can be the on-chain defects incorporated during synthesis. [48] Their oxidation leads to the presence of ketone defects, resulting in so-called fluorenone moieties incorporated in the polymer backbone. [49] In addition, this green emission may also be caused by interchain interaction. [50] It is important to note that, regardless of the mechanism responsible for this green emission such as interchain excited states or ketone defects, the functionalization of the polymer supramolecular organization can be exploited for the control of the solid-state photophysics. [50, 51]

Both EL and PL spectra for pure F8 based PLED have a common peak at ~ 441 nm, which is related to the β phase embedded in F8 amorphous phase matrix. [52, 53] For $F8_{0.95}SY_{0.05}$ based PLED, most of the emission is attributed to the SY (both in PL and EL). This devices exhibits its pure electronic transition peak (0-0 peak) at ~ 524 nm that undergoes a red-shift with increasing *x*. Pure SY PLED shows the 0-0 peak at ~551 nm (shifted ~ 27 nm relative to that of $x = 0.05$) and first vibronic peak (0-1 peak) at 601 nm.[54] The red-shift can be regarded as an evidence of SY aggregate formation and longer conjugation lengths with its increasing concentration. [55, 56] These aggreates possess low excicton energy because of the delocalisation of the electronic wavefunction among two or more chains. Consequently there is a red-shift interchain luminescence as compared with single chain exciton recombination. [57] Similar profiles of EL and PL spectra of pure SY PLED suggested that the emission sites are similar in origin. It is important to highlight that there is almost no F8 emission even for PLEDs with the lowest SY concentration ($x = 0.05$) in F8 matrix. We believe that there are two avenues by which F8 emission is quenched in as-made devices, namely a) trap assisted transport which is dominated by SY molecules, and b) Forster resonance energy transfer (FRET) which is favorable because of the strong overlap between photo-emission and photo-absorption spectra of F8 (host) and SY (guest) molecules, see **Supporting Information S2**. This paper deals only with the prior mechanism and later is the subject of another report.

The CIE (Commission Internationale de l'Eclairage) color coordinates relative to the EL spectra shows clear transition of the emission color from blue to yellowish green (/yellow) bypassing the white region when SY is added to F8. This shift of color is visible in the photographs of different devices, see **Figure 4** for CIE color coordinates and PLEDs in operation.

In order to understand the charge dynamics in as-made devices, single charge carrier devices provide vital information. Ca/Al cathode in PLEDs was replaced by MoO_3/Au anode to form the hole only devices (HODs). The device configuration (respective work-functions) for different layers are shown in **Figure 5**. MoO_3 is known to offer excellent Ohmic contact and efficient hole injecting layer for polymers having deep HOMO levels, such as F8 and F8BT. [19, 20, 41, 58, 59] Thermally deposited Au on MoO_3 works as electrical contact to subsequent MoO_3 layer.

Mott-Gurney space-charge-limited current (SCLC) equation combined with field dependent mobility can be used to calculate the hole mobility and helps in assessing the behavior of the bulk hole-conduction.[44] The charge transport is studied by fitting a simplified form of SCLC equation, enhanced by the Frenkel effect, namely

$$J_{SCLC} = 1.125[\varepsilon_0\varepsilon_r\mu_0(V-V_{bi})^2/d^3]\exp[\beta_F[(V-V_{bi})/d)]^{1/2}]$$

on the *J*-*V* curves of HODs when positively biased from MoO_3, where $\varepsilon_0\varepsilon_r$ is the dielectric constant of the polymer, μ_0 is the zero field mobility of holes, V is the applied voltage, $V_{bi} \approx 0.5$ V is the built-in potential, d is the thickness of the polymer layer, and β_F is the field-effect mobility coefficient related to the depths of the traps in the polymer.[19, 34, 43] Notice that hole-current density (J_h) reduces by 3 orders of magnitude upon mixing a very small amount of SY in F8 (see J_h-V curves for $x = 0$, and 0.05 at V ≈ 4.0 V). Curve fitting for J_h-V data of **Figure 5a** also gives the lowest value of $\mu_0 \approx 1.35\times10^{-9}$ $cm^2V^{-1}s^{-1}$ for $x = 0.05$, which is 3 orders of magnitude smaller than the highest value of $\mu_0 \approx 2.25\times10^{-6}$ $cm^2V^{-1}s^{-1}$ for $x = 0$.

A gradual increase in μ_0 is observed within $0.05 \leq x \leq 1$ for increasing value of *x*, which eventually approaches to $\sim 4.26\times10^{-7}$ $cm^2V^{-1}s^{-1}$ for $x = 1$, **Table 2**. In agreement to the behavior of μ_0, β_F shows the highest value of $\sim 8.05\times10^{-3}$ $cm^{1/2}V^{-1/2}$ for $x = 0.05$, which unsurprisingly decreases for the increasing value of *x*. We deduced that the reduced μ_0 is caused by introduction of SY traps in F8 which severely suppress the hole-current, especially in the case of $x = 0.05$. Severity of traps subsides when SY concentration iswas increased as holes begin to migrate by hopping mechanism between the SY traps due to their shrinking distance in F8 matrix. This explains the J_h-V trend and corresponding μ_0 (β_F) increasing (decreasing) behavior in $F8_{1-x}SY_x$ devices with increasing *x*.

Replacing the PEDOT:PSS layer in PLEDs with ZnO/Cs_2CO_3 layers gives an electron-only device (EOD). ZnO is generally used as electron injecting layer in LEDs.[19, 59] Cs_2CO_3 is also well-known for its electron injection properties.[20, 33, 41] The J_e-V curves for $F8_{1-x}SY_x$ based HODs with different values of *x* are shown in **Figure 5b**. The electron only devices do not fit well with SCLC and Pool-Frankel model, consistent with Kabra et al.[19] However, comparable current densities for hole only and electron only devices for $x = 0.05$ and 0.1 can be indicative of the fact that current density is bulk limited under this low charge density space-charge limited condition.[19] Pure F8 exhibits the highest electron current density $J_e \approx 13.3$ mA-cm^{-2} at $V \approx 4.0$ V, and it drops to ~ 4.2 mA-cm^{-2} for $x = 0.05$. Notice that this change in J_e becomes comparable to that observed in J_h for $x = 0.05$, an indicative for high performance at this composition. The decreasing trend of J_e persists throughout the concentration range $0 \leq x \leq 1$. This is because of the electron traps introduced by SY molecule, an intrinsic feature of PPVs.[60] The turn-on voltage (V_{on}) generally rises with further increase in *x*. Above turn-on we noticed a slower initial increase of J_e below a critical voltage, $V < V_c$ for $0.1 \leq x \leq 1$. We believe that the electrons transport is hindered because of increase in trap-density offered by the SY molecules for all $x > 0.05$. With increase in *x*, more

traps are introduced, pushing V_c further towards larger values. The applied potential above V_c starts liberating the trapped electrons from the trap sites, and they undergo a transition to SCLC behavior.[60]

A device with different electron and hole mobility causes the majority of the recombination to occur near the electrode surface.[19, 40, 46, 59] In such condition, the whole thickness of the emissive layer does not contribute in radiative recombination and cathode quenching comes into play leading to the poor device performance. If we can avoid this situation, a perfect charge balance between electron and hole transport exploits the whole thickness for emission and improves the efficiency. We believe that the blend PLEDs with $x = 0.05$, and 0.1 hold the benefit of fairly unmodified electron transport and these small SY concentration offer fewer traps compared with $x > 0.1$. However, hole-mobility is strongly suppressed for low SY concentrations because of the large difference between HOMO levels of both blended molecules: hole-traps offered by SY molecules reduce the current density by 3 orders of magnitude for $x = 0.05$ as compared with $x = 0$. Therefore, we conclude that it is the efficient charge balance in the $F8_{1-x}SY_x$ based PLEDs that causes the majority of exitonic recombinations within the bulk of the emissive layer, leading to a higher efficiency for such devices.

Note that SY is well-known for its high efficiency, ~ 14 cd-A^{-1}. An increase up to ~ 21 cd-A^{-1} in this report is a significant development. Kabara et al. have also achieved almost the same luminous efficiency for thick PLEDS, which is impressive as they have achieved these values in a non-blend PLED.[19] However, their devices require very large operating voltage. In contrast $F8_{1-x}SY_x$ based PLEDs are operated at low voltage and exhibit high power efficiency. We suggest that engineering of electrode-emissive layer interface would also increase the performance of these devices. Another optimization parameter is the thickness of the emissive layer. Since efficiency of $F8_{1-x}SY_x$ based devices is very high, the system would be

an interesting candidate for further exploration for injection lasing. Since, light emitting electrochemical cells are competing PLEDs and offer combatively low cost and easy processing, [36, 37, 39] incorporating $F8_{1-x}SY_x$ system into an electrochemical cell structure and tuning the device for high performance and stability will be an interesting study.

Summary

In summary, we propose a polymer blend, $F8_{1-x}SY_x$, that is based on mixture of poly (9,9-dioctylfluorene) (F8) and a poly (*para*-phenylenevinylene) (PPV) copolymer, superyellow (SY) as an efficient electro-limnescent system. The PLEDs based on this show excellent properties: luminance $(L) > 10^4$ cd-m^{-2}, luminous efficiency $(\eta) > 21$ cd-A^{-1} and, low operating voltage (V) ~ 3-10 V. The performance is because of the large difference (~ 0.6 eV) that exists between the highest occupied molecular orbital (HOMO) levels of F8 (~ 5.8 eV) and SY (~ 5.2 eV). In the mixture SY molecules slows down the hole and provide trap sites, pushing the excitonic recombination within the bulk of the material, away from the cathode. Thus a balanced charge injection into the emissive layer is responsible for overall increase in the device performance.

Experimental

Fabrication: (*Note*: Conditions for a particular layer were kept identical in all kinds of devices). Indium-tin-oxide (ITO) substrates were cleaned in acetone and isopropanol (10 minutes each) in an ultrasonic bath. Then, these substrates were treated with oxygen plasma. PEDOT:PSS was spin-coated and annealed at 230 ^{o}C for 30 minutes under nitrogen environment. Blend of F8 (294 kg-mol^{-1}, Cambridge Display Technology,) and SY (Merk, 616 kg-mol^{-1}) in chlorobenzene was spin-coated to obtain $F8_{1-x}SY_x$ films, and thickness of these films was controlled to around 100 ± 10 nm, confirmed by Dektak profilometer. These films were also coated on quartz substrates for optical measurements. After this deposition, the devices were annealed at 70 ^{o}C for 30 minutes in nitrogen. Ca/Al (3 nm/300 nm) electron

injection layer was thermally deposited in a vacuum chamber incorporated in a nitrogen glovebox. After completing the fabrication of active part and making the electrical conenections, devices were encapsulated for stable air operation. For hole only devices, MoO_3/Au (10 nm/70 nm) electrodes were made using thermal evaporation. For electron only devices, ZnO (50 nm) layer was deposited by spray pyrolysis on heated ITO substrate at 370 ^oC. Organic precursor zinc acetate dihydrate (Fluka) dissolved in anhydrous methanol (80 g-L^{-1}) was used for this deposition. Cs_2CO_3 (Sigma Alrich) was dissolved in 2-methoxyethanol (5 g-L^{-1}, Fluka) and spin-coated on ZnO layer. A spin speed of 6000 rpm resulted 2 nm thick Cs_2CO_3 layers.

Characterization: Current density-voltage-luminance (*J-V-L*) measurments of PLEDs were performed in air using computer contolled sourcemeter (Keithley 2400) and chromameter (Minolta CS-200). Electroluminescence (EL) spectra was measured with a Ocean Optics spectrometer (USB2000). For photolumenscence, the polymer films were optically excited with 3-5 ns (10 Hz) laser pulses obtained from a compact housing of the nanosceond optical parameteric oscillator (OPO) system and Nd:YAG Q-switched laser (NT342B, EDSPLA). The spectra was taken using 500 mm spectrograph, (SpectraPro2500i, Princeton Instruments) with incorporated CCD camera (Pix 100-F, Princeton Instruments).

Acknowledgements

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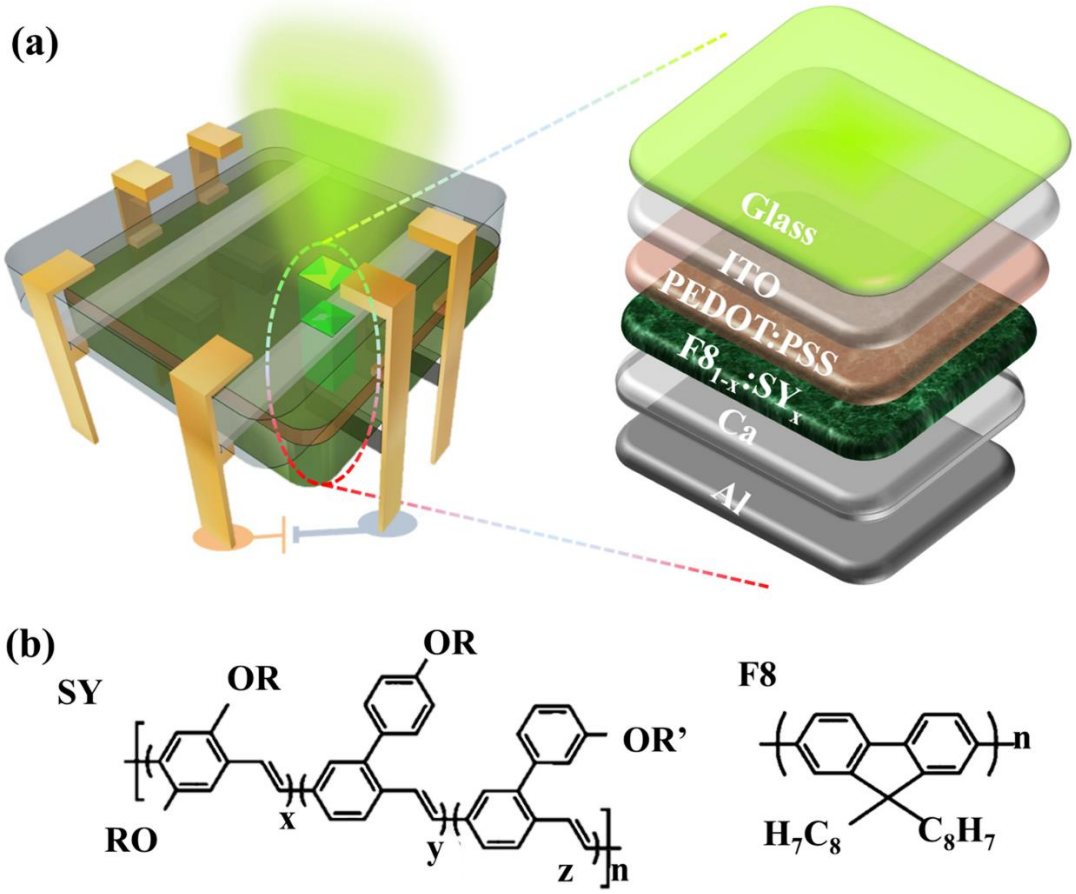

Figure 1. a) Construction of $F8_{1-x}SY_x$ based polymer light emitting diode (PLED) pixels and different layers comprising the pixel, namely Glass/ITO/PEDOT:PSS/$F8_{1-x}SY_x$/Ca/Al. **b)** Chemical structures of F8 and SY molecules.

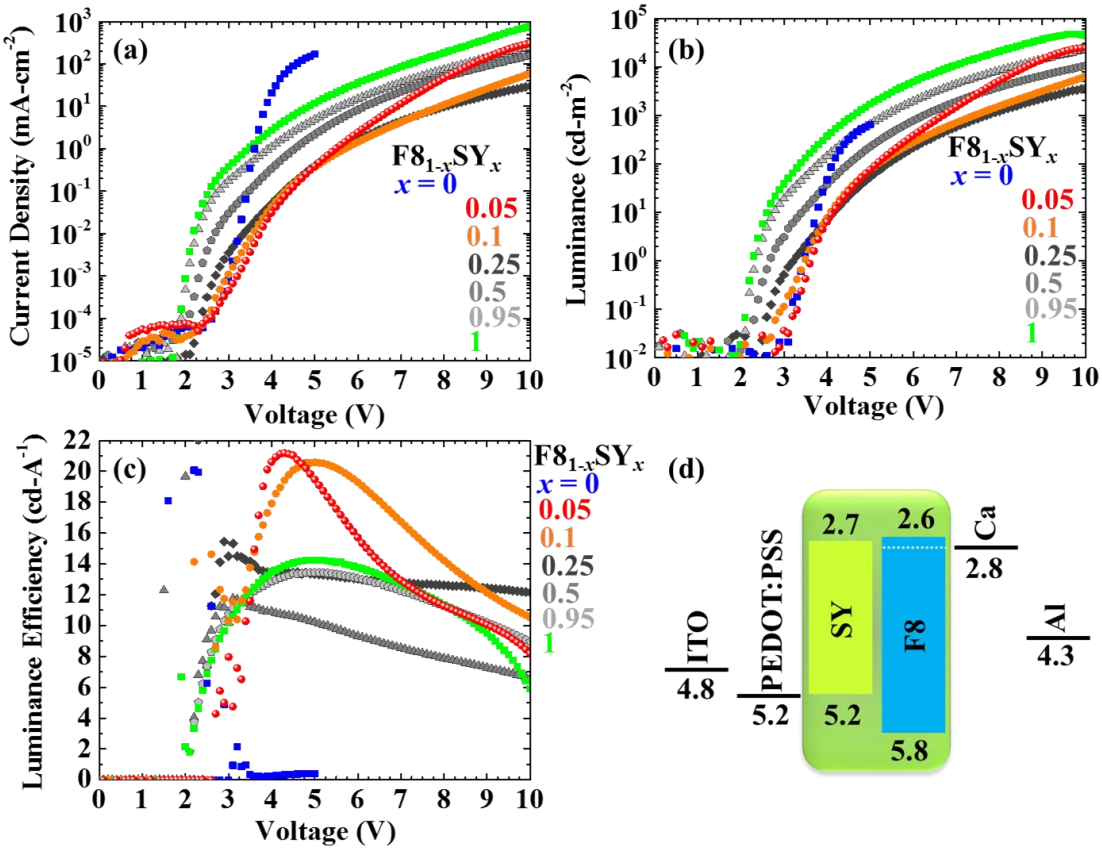

Figure 2. a) *J-V* curves of $F8_{1-x}SY_x$ based PLEDs with increasing SY weight concentration *x*: The blend system exhibits a drastic decrease in current density for $x = 0.05$ and 0.1 because of hole traps offered by SY molecules – holes move via a trap-detrap mechanism. Subsequently, a hopping mechanism begins to dominate the hole transport as SY come closer with increasing *x*. **b)** *L-V* curves: luminance almost shows similar trend as *J-V*. However, as compared with *J-V* light turns on at a larger voltages for all values of *x*. Maximum *L* is exhibited by pure SY LED, whereas that with x = 0.05 initially shows a late start but approaches the value of pure LED as $V \rightarrow 10$ V. **c)** η-*V* curves for $F8_{1-x}SY_x$ based PLEDs: First (second) highest luminous efficiency of ~ 21.2 (20) cd-A^{-1} is achieved for $x = 0.05$ (0.1) which is significantly larger than the pure SY PLEDs ($\eta \approx 14$ cd-A^{-1}), see **Table 1** for more details. **d)** Energy band diagram of each layer in $F8_{1-x}SY_x$ based PLED. The blue dash line in the F8 shows its β-phase LUMO level which is slightly lower than the amorphous phase. The

large difference between the HOMOs of both molecules causes hole traps severely reducing the hole mobility.

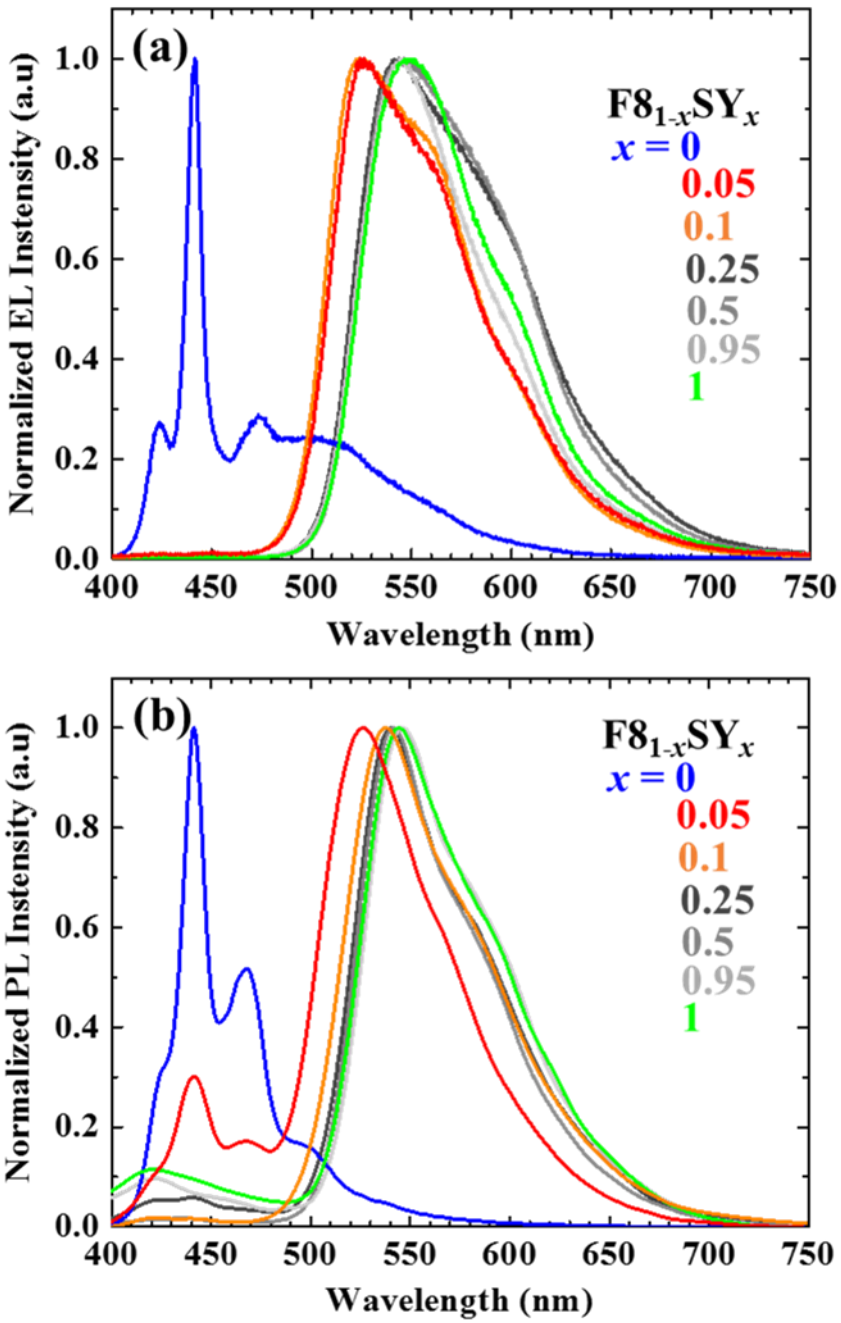

Figure 3. a) Normalized EL spectra of the $F8_{1-x}SY_x$ based PLEDs with increasing SY concentrations taken at 5 V. Notice the strong suppression of F8 spectra for only a very small of SY x = 0.05, indicating that most of the recombination is occurring on SY sites. The range $0.1 < x < 0.95$ shows a wavelength broadening. **b)** Normalized PL spectra of $F8_{1-x}SY_x$ blend system also shows a strong suppression of F8 spectra upon small mixing of SY. This is because of efficient Förster resonance energy transfer between F8 and SY molecules.

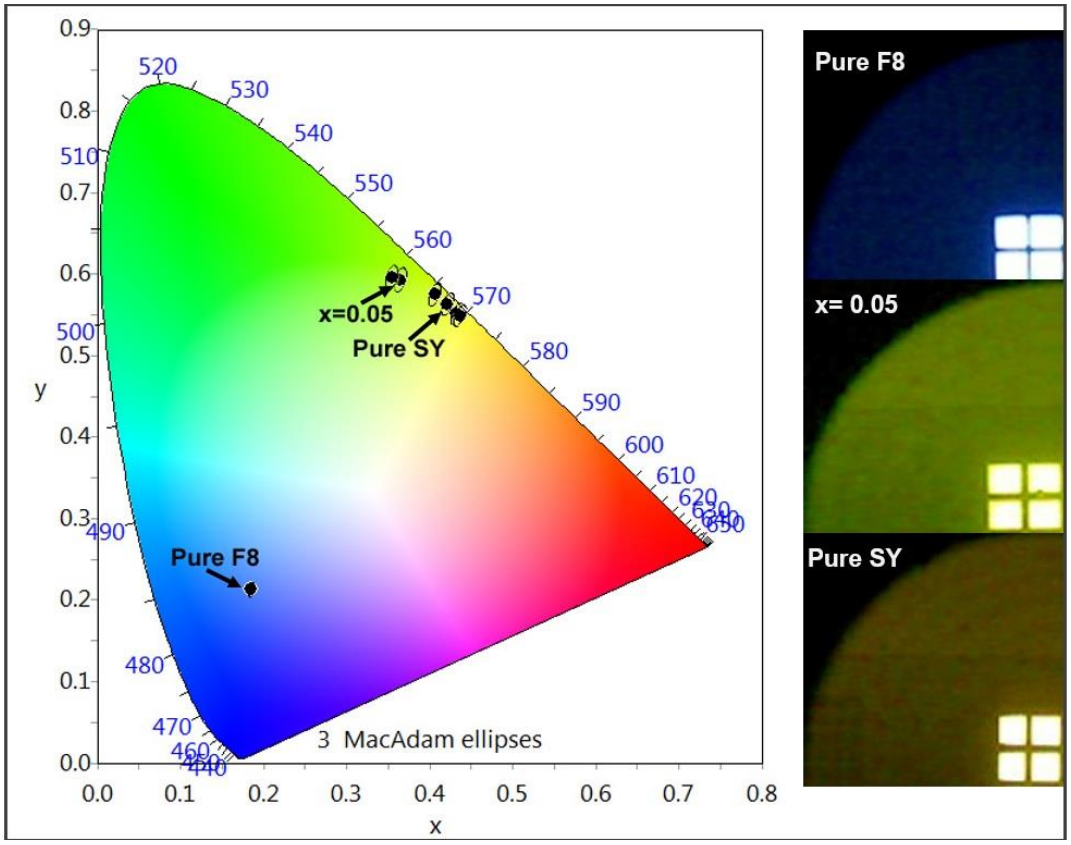

Figure 4. The CIE color coordinates relative to the EL spectra (left) and photograph of three corresponding devices with pure F8, $F8_{0.95}SY_{0.05}$ and pure SY emissive layers (right). F8 devices emit in blue region which changes to yellowish green upon small mixing with SY. All devices with some SY concentration emit near to this green/yellow region.

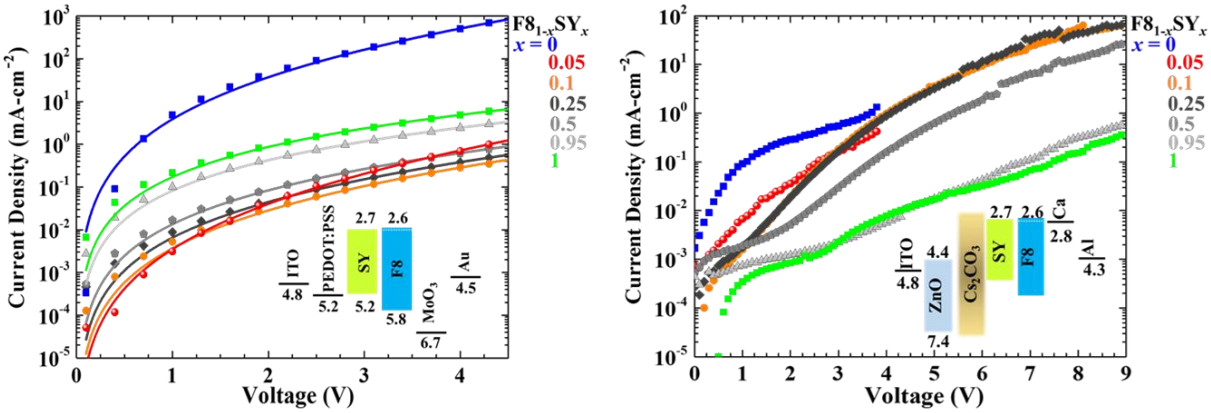

Figure 5. a) *J-V* curves for $F8_{1-x}SY_x$ based hole only devices with different value of *x*. The devices are positively biased from the MoO_3 side. The fitting results are shown with solid lines for each curve (see **Table 2**). The current is severely reduced for $x = 0.05$ and 0.1 indicating hole traps offered by SY molecules. This is because of the large difference present between the HOMOs of F8 and SY. Inset shows the energy band diagram of each layer in the hole only device (Glass/ITO/PEDOT:PSS/$F8_{1-x}SY_x$/MoO_3/Au). The blue dash line in the F8 depicts its *β*-phase LUMO level. **b)** *J-V* curves for $F8_{1-x}SY_x$ based electron only devices with increasing SY concentration: The devices are negatively biased from the Ca/Al electrode. There is reduction in electron current for x = 0.05 as compared with pure F8 devices, however at much smaller scale as compared with reduction observed in hole current in hole only devices. Inset shows the energy band diagram of each layer in the electron only device (with schematic Glass/ITO/ZnO/Cs_2CO_3/$F8_{1-x}SY_x$/Ca/Al).

Tables.

SY wt. x in $F8_{1-x}SY_x$	L (cd/m^2)	η_{max} (cd/A)	V_{on} (V)	PE_{max} (lm/W)	EQE_{max} (%)
0 (pure F8)	634 @ 5.0 V (max.value)	0.9 @ 3.4 V	2.4	0.07[40]	0.2 [46] 0.69[47]
0.05	1000 @ 6.7 V	21.2 @ 4.4 V	2.3	8	6.9
0.1	1000 @ 7.4 V	20.6 @ 5.0 V	2.3	8.2	7.3
0.25	1000 @ 7.7 V	15.4 @ 2.9 V	2.1		
0.5	1000 @ 6.2 V	15.0 @ 4.0 V	1.8		
0.95	1000 @ 5.3 V	13.4 @ 4.9 V	1.7		
1 (pure SY)	1000 @ 4.6 V	14.2 @ 5.0 V	1.6	6[37]	3.36[47]

Table 1. Summary of some performance parameters of $F8_{1-x}SY_x$ based PLEDs.

SY wt. x in $F8_{1-x}SY_x$	μ_0 ($cm^2V^{-1}s^{-1}$)	β ($cm^{1/2}V^{-1/2}$)
0 (pure F8)	2.25×10^{-6}	6.70×10^{-3}
5	1.35×10^{-9}	8.05×10^{-3}
10	4.22×10^{-9}	4.70×10^{-3}
25	7.94×10^{-9}	4.17×10^{-3}
50	2.06×10^{-8}	3.41×10^{-3}
95	1.96×10^{-7}	1.97×10^{-3}
100 (pure SY)	4.26×10^{-7}	1.87×10^{-3}

Table 2. Hole transport parameters extracted by fitting hole only current injected from MoO_3/Au electrodes.

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